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THERMAL PROPERTIES OF NON-CRYSTALLINE SOLIDS HAVING MAJOR CONTRIBUTION OF TWO LEVEL SYSTEM

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Abstract : The aim of the present work is to develop a self consistent approach to the two level system (TLS) of glassy material at very low temperature. Hence we shall consider those properties of glassy substances which depend solely on proposed TLS's present in glass. Hence we shall limit ourselves only to very low temperatures in which TLS effect predominates. Two level system model successfully explains the anomalies but subsequently has put forward a large task in the completeness. Here a theoretical explanation of change of Gruneisen parameter with temperature has been attempted successfully as was shown in tabular form at the end from measurements of Ackerman.

Keywords : Gruneisen parameter, TLS.

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1. INTRODUCTION

Before seventies it was thought that glasses having no specific structure will allow only low q phonons to pass through them and hence behave like a continuum to elastic waves. Naturally no significant changes in their behaviour from their crystalline counterpart was expected although experiments claiming otherwise. Indication of presence of some unknown excitation in amorphous solids came from the measurements of thermal properties [1,2]. Zeller and Pohl [3] measured specific heat and thermal conductivity of a number of glasses accurately up to 0.2K. Stephen [4,5] extended these experiments down to 0.05K and to several other glasses. Thermal expansion was first studied by White and till date the magnitude of Gruneisen paramenter remains a mystery. These experiments indicate that specific heat was fairly independent of silica sample. But the work by Lasjaunias [6,7] et al which was extended to 0.025K demonstrated the sample dependence on behaviour. Lasjaunias [7] et al suggested the steeper temperature dependence resulted from a gap in the low energy density of states. But in this model magnitude of Gruneisen

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parameter varies as T^{-2} which supports measurements of Ackerman [8] et al. In a perfect crystal atoms occupy the single potential minima. But when impurities are introduced the defects or the impurity atoms move in multi minima potential provided by neighboring atoms. At low temperature the tunneling of the atoms between the two minia gives rise to a vey small energy splitting of the ground state. According to this model certain atoms or groups can occupy two or more potential minima of nearly equal energy. The hypothesis is consistent with observed increase of entropy [9, 10] of glass with respect to crystal. "Since temperature is very small changes in energy will certainly be very small. Hence due to small energy splitting of ground state we can regard the tunneling entity just like a free particle of momentum p and energy $p^2 / 2M$. Hence we can say density of states proportional to $E^{1/2}$, which is also supported by Lasjaunias. In spectrosil-W density of states at 1K is 9×10^{32} / erg-cc and changes to 3×10^{32} / erg-cc at 0.1 K. Here tunneling of entities explicitly assumes a double well potential. If Δ be the difference in the two potential minima and $\Delta_0 = \hbar\omega_0 \exp [-d (2mv)^{1/2} / \hbar]$ where $\hbar\omega_0$ is the zero point energy in either well, V is the potential barrier, d is the distance in the configuration space between the two potential minima, and m is the mass of the tunneling entity [11]. Properties calulated with this model will be highly sensitive to the form of distribution function $f (\Delta, \Delta_0)$ for Δ and Δ_0.

2.1. CALCULATION OF SPECIFIC HEAT, GRUNEISEN PARAMETER

We know free energy, $F (E) = - k_b T \ln [2 \cosh (E / 2k_b T)]$.

So contribution to specific heat is readily found to be

$$C(E) = (E^2 / 4k_b T^2) \operatorname{sech}^2 (E / 2k_b T) , \qquad (1)$$

$$C = A \int_0^\infty C (E)\, p(E)\, E^{1/2}\, dE . \qquad (2)$$

where, $p (E) = [1-2(e^{E/k_b T}+1)^{-1}]$ is the occupation probability of ground state of TLS and $AE^{1/2}$ is the density of states of TLS, where A is a constant. Hence

$$C = Ak_b^{5/2} T^{3/2} \Gamma(7/2)\, \eta\, (3/2) \qquad (3)$$

Thus specific heat is given by $c = a_1 T^{3/2} + b_1 T^3$ (4)

Entropy of a single TLS can be written in the form $S = - \left(\frac{\partial F}{\partial T}\right)_v$ (5)

From thermodynamics volume expansion coefficient α and isothermal compressibility χ of a single TLS is given by, $\alpha = \chi(\delta S / \delta v)_T$.

The contribution to Gruneisen parameter Ω to TLS is

$$d\Omega = -\frac{E \operatorname{sech}^2 (E/2k_b T)(D_l + 2D_t)}{2\pi^2 n_0 k_b^3 T^3} , \qquad (6)$$

D_l & D_t are longitudinal and transverse deformation potential repectively.

Multiplying (6) by (2) and integrating we get

$$\Omega C = 2A \frac{(D_l + 2D_t)\,\Gamma(9/2)}{\pi^2 n_0 k_b^{1/2} T^{1/2}} \zeta(9/2)\,. \tag{7}$$

Thus we have $\Omega T^2 / (D_l + 2D_t)$ = constant

2.2. CALCULATION OF THERMAL CONDUCTIVITY

In analogy with results of standard kinetic theory of gas we can write the phonon contribution to conductivity of a crystalline medium as

$$K(T) = \frac{1}{3}\sum_{\alpha} \int_0^{\omega_D} C_\alpha(\omega) V_\alpha(\omega)\, l_\alpha(\omega)$$

$$C_\alpha(\omega) = \left(\frac{\hbar\omega}{2k_bT}\right)^2 \operatorname{sech}^2\left(\frac{\hbar\omega}{2k_bT}\right) \frac{\omega^2}{\quad} k_b\,,$$

where l_α of phonons is related to its transition probability per unit time τ^{-1} in the resonant absorption process. For calculation of transition probability we require the transition matrix element $<2|H|1> = (\omega/2\rho V^2)^{1/2} M_\alpha$. Where M_α is the coupling constant of these phonons. Thus the transition probability per unit time can be calculated from the Golden rule [14].

$$\tau^{-1} = \frac{2\pi}{h} n <2|H|1> [1-(e^{\hbar\omega/k_bT}+1)^{-1}] \qquad \text{Hence}$$

$$K(T) = k_b^3 \sum \frac{v_\alpha \rho T^2}{M_\alpha^2 \pi^3 \hbar^2}\, \eta(3)\Gamma(4) = DT^2\,. \tag{8}$$

Thus we have $\dfrac{K(T)}{C(T)}$ = constant $(T^{1/2})$

3. DISCUSSION

According to Lasjaunias $C = c_1 T^n + c_D T^3$, where n = 1.5 for B_2O_3 and 1.3 for SiO_2. In earlier models deformation potential was taken as constants. Deformation potential should be energy dependent as remarded by Ackerman, hence we find temperature dependence of Ω and it is also reflected in the measurements of Ackerman [8]. We use formula

$$C = C_{TLS} + C_{DEBYE} = a_1 T^{3/2} + b_1 T^3 \tag{9}$$

Another contribution of this model is that it is forecasting values of Ω from the

average values of $\Omega T^2 / \alpha$ which was shown in this model as constant. Also in the tables, it is shown that $\Omega T^2 / \alpha$ is fairly constant within limitation of measurements. Asterisk (*) provided some postulated values of Ω which is yet to be confirmed by experiments.

TABLE 1 As_2S_3 $a_1 = 0.2 J/cm^3K^{5/2}$, $b_1 = 3.5 J/cm^3K^4$, $\delta = 0.4$

T	$10^5 C/T$	$10^{10}\alpha/T$	$10^5 C/T^{3/2}$	$10^5 C_{TLS}$	$10^4 K$	Ω	$\Omega T^2/\alpha$	K/CT^δ
K	J/cm^3K^2	K^{-2}	$J/cm^3K^{5/2}$	J/cm^3K	W/cmK		$10^{10}K^3$	$10cm^2/sK^\delta$
0.1	0.136	-0.41	0.430	0.0063	0.2044	-0.58*		81.23
0.2	0.244	-0.14	0.546	0.0178	0.7732	-0.10*		80.73
0.3	0.424	0.31	0.774	0.0329	1.658	0.15*		82.79
0.4	0.676	0.94	1.069	0.0506	2.927	0.34*		83.46
0.5	1.000	1.75	1.400	0.0707	4.492	0.51*		83.83
0.6	1.396	2.74	1.800	0.0929	6.375	0.75	0.163	84.14
0.7	1.864	3.91	2.227	0.1171	8.571	0.85	0.152	84.42
0.8	2.404	5.26	2.680	0.1431	11.06	0.93	0.141	84.51
0.9	3.016	6.79	3.178	0.1708	13.87	1.0	0.133	84.70
1.0	3.700	8.50	3.700	0.2000	17.00	1.2	0.141	85.00

Average value of $\Omega T^2 / \alpha = 0.156 \times 10^{10} K^3$

TABLE 2 Spectrosil-B $a_1 = 0.4 J/cm^3K^{5/2}$, $b_1 = 0.31 J/cm^3K^4$, $\delta = 0.5$

T	$10^5 C/T$	$10^{10}\alpha/T$	$10^5 C/T^{3/2}$	$10^5 C_{TLS}$	$10^4 K$	Ω	$\Omega T^2/\alpha$	K/CT^δ
K	J/cm^3K^2	K^{-2}	$J/cm^3K^{5/2}$	J/cm^3K	W/cmK		$10^{10}K^3$	$10cm^2/sK^\delta$
0.1	0.2546	-8.025	0.8057	0.02515	0.0324	-136*		3.23
0.2	0.2684	-8.100	0.6004	0.05120	0.1183	-68*		4.42
0.3	0.2914	-8.225	0.5318	0.07905	0.2526	-45.4*		5.17
0.4	0.3236	-8.400	0.5116	0.10960	0.4236	-34*		5.69
0.5	0.3650	-8.625	0.5163	0.14375	0.6566	-27	1.69	6.02
0.6	0.4156	-8.900	0.5360	0.18240	0.9233	-23	1.73	6.21
0.7	0.4754	-9.225	0.5680	0.22645	1.2320	-19	1.67	6.35
0.8	0.5444	-9.600	0.6090	0.27680	1.5810	-17	1.70	6.25
0.9	0.6262	-10.025	0.6600	0.33759	1.9710	-16	1.80	6.09
1.0	0.7100	-12.500	0.7100	0.40000	2.4000	-15	1.87	6.00

Average value of $\Omega T^2 / \alpha = 1.73 \times 10^{10} K^3$

TABLE 3 PMMA $a_1 = 1.2 J/cm^3K^{5/2}$, $b_1 = 2.47 J/cm^3K^4$, $\delta = 0.4$

T	$10^5 C/T$	$10^{10}\alpha/T$	$10^5 C/T^{3/2}$	$10^5 C_{TLS}$	$10^4 K$	Ω	$\Omega T^2/\alpha$	K/CT^δ
K	J/cm^3K^2	K^{-2}	$J/cm^3K^{5/2}$	J/cm^3K	W/cmK		$10^{10}K^3$	$10cm^2/sK^\delta$
0.1	0.700	-2.89	2.215	0.06753	0.0511	-1.76*		1.90
0.2	0.790	-2.56	1.767	0.13824	0.1792	-0.79*		1.90
0.3	0.940	-2.01	1.715	0.21531	0.3733	-0.41*		2.81
0.4	1.150	-1.24	1.818	0.30192	0.6284	-0.19*		3.00
0.5	1.420	-0.25	2.008	0.40125	0.9411	-0.03*		3.09
0.6	1.750	0.96	2.250	0.51648	1.3090	0.1	0.062	3.11
0.7	2.140	2.39	2.557	0.65079	1.7300	0.2	0.059	3.07
0.8	2.590	4.04	2.896	0.80736	2.2.30	0.3	0.060	2.98
0.9	3.100	5.91	3.440	0.98937	2.7270	0.4	0.061	2.87
1.0	3.670	8.00	3.670	1.20000	3.3000	0.5	0.062	2.75

Average value of $\Omega T^2 / \alpha = 0.061 \times 10^{10} K^3$

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